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13. ABSTRACT *(Maximum 200 words)*

Significant advances were made in the technology of molecular tagging diagnostics. Taking advantage of recently engineered water-soluble molecular complexes with long-lived excited states (i.e. phosphorescence), techniques were developed for non-intrusive, multi-point, measurements of various fluid dynamical quantities. As a result of two novel innovations, the single-point accuracy of velocity measurements was improved by about a factor of 5, while the density of measurement points in a plane was increased by an order of magnitude. It was demonstrated that the same chemical compound used for velocimetry could be utilized to molecularly tag either a chemical reaction interface between two streams, or a passive scalar (non-reacting) mixing region, and monitor the Lagrangian evolution of the tagged regions. This development offers the capability for simultaneous velocity and scalar mixing measurements. The technique has now reached a sufficient level of maturity that it can be applied to the study of various flows.

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Novel Diagnostics for the Study of Mixing and Chemical Reactions in Turbulent Liquid Phase Flows

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1. Introduction

Laser induced fluorescence (LIF) diagnostics have developed into a most powerful tool for studying scalar mixing in liquid-phase flows. In most cases, LIF data are not, by themselves, sufficient for gaining a fundamental understanding of entrainment and mixing dynamics. Additional flow kinematics (i.e. velocity, vorticity) are needed for this purpose. This provides the motivation for developing "molecular tagging diagnostics" under this AASERT grant in order to supplement our LIF capabilities. The initial beneficiary of these developments is the parent grant F49620-93-1-0282, where we are investigating the influence of 2-D and 3-D disturbances on scalar mixing in shear layers and wakes [1-5].

As a background, it is worth noting that fluorescence is a short-lived excited state, typically with a lifetime of a few nanoseconds. Therefore, fluorescent molecules are generally not suitable for obtaining flow kinematics information. Our developments rely on recently engineered water-soluble molecular complexes with long-lived excited states (i.e. phosphorescence). The molecular nature of the diagnostics offers the potential for simultaneous velocity/scalar measurements, a task nearly impossible to accomplish with particle-based techniques such as LDV or PIV.

Work under this grant concentrated on the development of new capabilities and refinement of the various aspects of molecular tagging diagnostics to provide a powerful tool for research. We believe that the technique has now reached a sufficient level of maturity that it can be applied to the study of various flows. This report provides a summary of our progress over the last three years. The support provided by this grant has been instrumental in the education and research training of a number of undergraduate and graduate students. This aspect of the grant is also described.

2. Molecular Tagging Diagnostics

Molecular tagging diagnostics take advantage of molecules with long-lived excited states for non-intrusive, multi-point, measurements of various fluid dynamical quantities. The technique relies on the capability to tag portions of the flow by a laser and monitor their subsequent evolution over the luminescence lifetime of the molecule. We have significantly advanced this technique beyond its original form devised by R. E. Falco at MSU for the purpose of velocimetry [6]. Upon implementing a series of innovations (see Section 2.1), the single-point accuracy of velocity measurements has improved by about a factor of 5, while the density of measurement points in a plane has increased by an order of magnitude. Even more importantly, we have been able to demonstrate that the same chemical compound used for velocimetry can be utilized to molecularly tag either a chemical reaction interface between two

streams, or a passive scalar (non-reacting) mixing region, and monitor the Lagrangian evolution of the tagged regions. This development naturally leads to the capability for simultaneous velocity and scalar mixing measurements.

Our developments to date have centered around a triplex phosphorescent compound, alcohol/CD cup/lumophore, developed by Nocera's group at MSU's Chemistry Department [7]. This compound is water soluble and has a lifetime $\tau \approx 5$ ms. We have devised three different implementations depending on the placement in the flow of the three components of the compound [8-9]. Details are given below.

2.1 Molecular Tagging Velocimetry (MTV)

When the three components (i.e. alcohol, CD cup, and lumophore) are premixed with water in the entire flow facility, we obtain a technique intended solely for velocity measurements. A pulsed laser is used to molecularly tag the regions of interest. The tagged regions are imaged at two successive times within the phosphorescence lifetime. The measured Lagrangian displacement vector provides the estimate of the velocity vector. To measure two components of the velocity in a plane, the luminescence intensity field must have spatial gradients along two, preferably orthogonal, directions. For single-point velocimetry, this is easily achieved using a pair of crossing laser beams; a grid of intersecting laser lines allows multi-point velocity measurements [6,8,9].

The accuracy of velocity measurements is dictated by the accuracy with which the displacement vector is determined. Therefore, every attempt should be made to achieve the highest sub-pixel accuracy in displacement measurement. We have made significant improvements in the accuracy of the MTV technique by using two-detector imaging and measuring the displacement of the tagged regions by direct digital spatial correlation techniques. The experimental arrangement, shown in Figure 1, involves a link between the pulsed laser and two image detectors through a digital delay generator. Immediately after the laser firing, the first detector records the first image, and after a prescribed time delay Δt, the second detector records the second image. These images are acquired by two separate acquisition systems connected to the two detectors. The advantage of this arrangement is that any spatial "wandering" of the tagged regions (e.g. due to laser beam pointing instability or flow facility vibration) does not contribute to error in the measurement of displacement of these regions.

The displacement of the tagged regions is determined using a direct digital spatial correlation technique. The example provided in Figure 2(a) illustrates a computer simulation of regions tagged by laser grid at two different times and the corresponding spatial correlation coefficient field for one grid crossing. A well-defined correlation peak occurs at the location corresponding to the displacement; the peak is located using a multi-dimensional polynomial fit. A major advantage of our processing technique over traditional methods, which locate the centers of the crossing laser beams, is robustness to the presence of noise. This is due to the

averaging process inherent in the correlation procedure. The example in Figure 2(b) shows that a well-defined correlation field is obtained even with images of poor signal to noise ratio (S/N). An example of a grid crossing (at two different times) from an experiment, and the related correlation field is shown in Figure 3. We have found, based on both experiments and an extensive set of simulations, that we can measure the displacement of the tagged regions with ±0.1 sub-pixel accuracy (95% confidence limit). This is at least a factor of 5 improvement over previous approaches. We believe refinements in the experimental setup and data processing can lead to more improvements.

An example of the quality of data obtained using MTV is shown in Figure 4. This figure illustrates the time evolution of two components of the velocity vector near the axis of symmetry of a passing vortex ring. We note that this represents raw data (i.e. no filtering/smoothing) from one of 77 grid points where simultaneous velocity data were obtained.

Work has recently begun to apply MTV to the highly 3-D flow field of a forced wake. We have previously reported that forcing a low Reynolds number 2-D wake can lead to a highly three-dimensional flow and a large increase in mixing [3,5]. Preliminary instantaneous velocity vectors (v, w components) in the cross-stream (y-z) plane at downstream station x = 13 cm are shown in Figure 5. The region shown covers the central 2 cm × 3.7 cm of the test section cross sectional area. Except for remapping onto a regular grid, these are raw data; no attempt was made to remove the "bad" vectors or apply spatial filtering. Note that the mean streamwise flow direction is out of the page. In comparing the unforced wake, Figure 5(a), with the forced wake, Figure 5(b), it is clear that forcing has lead to a tremendous increase in three-dimensionality and streamwise vorticity. The maximum velocity in Figure 5(b) is about 60% of the wake freestream velocity.

2.2 Passive Scalar Mixing Dynamics

In the non-reacting (passive scalar) implementation, the alcohol/CD cup/lumophore are premixed in one stream and the other mixing stream is either pure water or contains an alcohol solution. The Lagrangian evolution of the scalar mixing field is then monitored over the luminescence lifetime. An example of this implementation is shown in Figure 6. A vortex ring fluid is premixed with the alcohol/CD cup/lumophore solution, and the ambient fluid contains only the alcohol solution. The UV laser was arranged to illuminate a series of parallel "bands" in the flow, and the resulting luminescence was imaged simultaneously on two CCD cameras with a prescribed time delay between them. Figure 6 shows the molecularly-tagged patches of the vortex ring fluid at the initial time (20 μs after laser firing), marked by the light shade of grey, and the evolution of the same patches 9 ms later (dark shade of grey). The velocity vectors at the corners of the patches, determined by the same spatial correlation technique used in MTV. The velocity vectors in the laboratory frame and the vortex frame are illustrated in Figures 7 and 8.

Even though the vortex ring example just shown has a rather simple concentration field (i.e. uniform within the tagged patches), it does highlight the potential of a molecular tagging approach for simultaneous concentration (from the first zero time delay image) and velocity (using image pairs as in Figure 6) measurements in more complex and turbulent flows. In this case, the spatially non-uniform scalar concentration field typical of most turbulent flows is used as a natural source of the luminescence intensity variation described earlier. Where this can be accomplished, a much simpler optical arrangement with a laser sheet replaces a more involved technique of "writing" a prescribed laser pattern into the flow (e.g. the usual grid pattern). Preliminary results illustrating this approach are discussed below.

Figure 9 is an image of the passive scalar field in a downward moving turbulent vortex ring recorded 1 μs after being tagged by two crossing laser sheets. The horizontal and vertical extents correspond to 53 mm and 71 mm, respectively. We demonstrate the use of the spatial correlation technique for the extraction of the displacement field for the three sample windows marked in Figure 9. The enlarged views showing the intensity distribution within these three sample windows, the image of the same tagged molecules 6 ms later, and the corresponding spatial correlation coefficient for each case are indicated. We note a well-defined correlation peak in cases 1 and 2 where there is sufficient intensity gradients (in two directions) within the chosen windows. The two components of the velocity vector can be determined in these cases. In contrast, the correlation field for window 3 allows the determination of the displacement only along one direction (normal to the interface shown); the absence of a "peak" in the orthogonal direction is connected to the lack of intensity variation along the interface.

Figure 10 shows the raw velocity map for the flow in Figure 9 determined by the procedure just described. The maximum velocity is about 23 cm/s and the sample window used for the correlation was a square 21 pixels on the side corresponding to about 3.6 mm in size. It is very important to appreciate that this raw velocity field is our zeroth attempt. We simply fed the two images to our correlation program using a fixed correlation window size and obtained the vector field shown; no attempt was made to pre-process, post-process, or optimize the technique. There are obvious areas where the velocity vectors do not "look" right; these are mostly due to low signal/noise, no signal, small spatial features of the flow compared to the fixed correlation window size used, or lack of a sufficient scalar gradient in a second direction. Many of these issues can be resolved by optimizing our processing and, if needed, using a non-uniform laser illumination (grids being a special case) to tag the scalar field.

2.3 Chemically-Reacting Interface Dynamics

In the chemical-reaction implementation of this technique, two freestream fluids are prepared by premixing the alcohol solution in one and the CD cup/lumophore in the other. The phosphorescent complex is formed only where the two freestream fluids are molecularly mixed. The Lagrangian evolution of the reaction interface is then monitored over the

luminescence lifetime. An example is shown in Figure 11(a) illustrating the reaction interface between a vortex ring fluid and ambient fluid imaged initially at 20 μs after tagging by a laser sheet. Figure 11(b) shows the tagged interfaces 6 ms later. Figure 11(b), which was obtained after significant digital image enhancement, highlights the inadequacy of our current non-intensified CCD arrays for this applications. As a result, we are not pursuing the chemically reacting flows any further until we get access to appropriate intensified detectors.

3. Education and Training of Students

Over the last three years, this AASERT program has provided either full or partial support for the education and training of 2 graduate and 3 undergraduate students (all U.S. citizens). Two of these undergraduate students are continuing their education and research in PhD programs. A brief summary of each student's involvement is provided below.

1. Charles Gendrich (graduate student)

Mr. Gendrich has been the mastermind behind our molecular tagging developments to date. This work is part of his PhD thesis research which is expected to complete in about a year. Mr. Gendrich is one of our most qualified students; typically in the upper 2% of his class and with a current GPA of 3.82/4.0, he received in 1994 an academic award as the top student in the Mechanical Engineering Department at MSU. He has been a past recipient of an AFOSR graduate fellowship.

2. Richard Cohn (graduate student)

Mr. Cohn has been involved with this AASERT grant for a year. As part of his PhD thesis research, he is applying molecular tagging diagnostics to highly 3-D flows in forced shear layers and concentrated vortices. Mr. Cohn is a past recipient of a Churchill fellowship (one of ten awarded nationally), and currently holds an NSF graduate fellowship.

3. Amy Warncke (undergraduate student)

Miss Warncke worked on the initial developments regarding the hardware and software integration of our two-detector imaging system. She graduated with a GPA of 3.95/4.0 and received an NSF graduate fellowship. She is currently pursuing her PhD degree in the Aeronautics Department at Caltech.

4. Steven Beresh (undergraduate student)

Mr. Beresh worked on the development and application of scientific visualization tools to analyze our large volumetric data sets. He graduated with a GPA of 3.95/4.0 and is now

working towards a PhD degree at the University of Texas in Austin in the area of high-speed compressible shear layers.

5. Michael Tsai (undergraduate student)

Mr. Tsai spent a summer working on the applications of our spatial correlation processing technique to various data sets. He is now working as an engineer at an auto company in Michigan.

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9. Gendrich, C. P., Koochesfahani, M. M. and Nocera, D. G. [1994] *Bull. Am. Phys. Soc.*, **39(9)**, 1980.

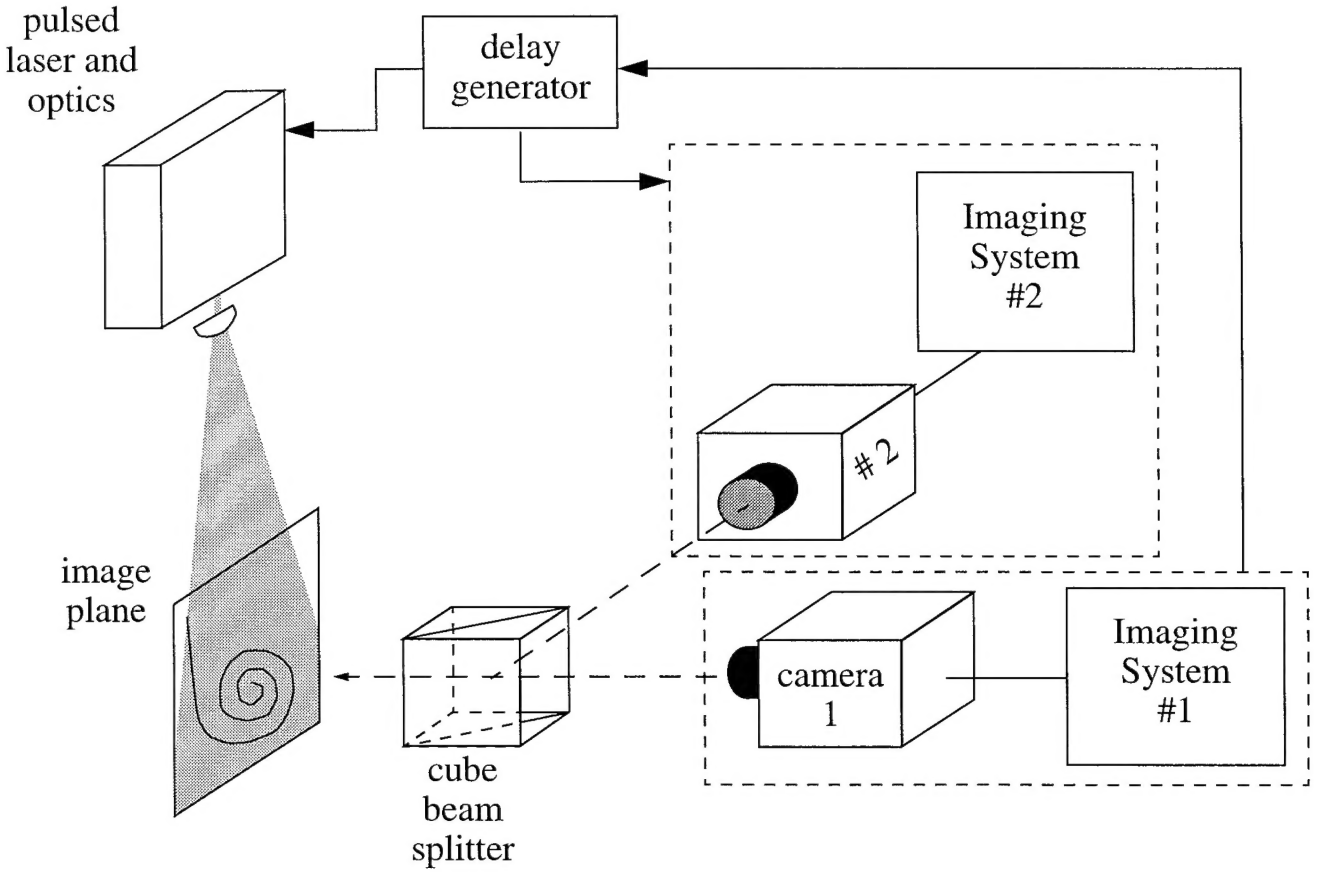
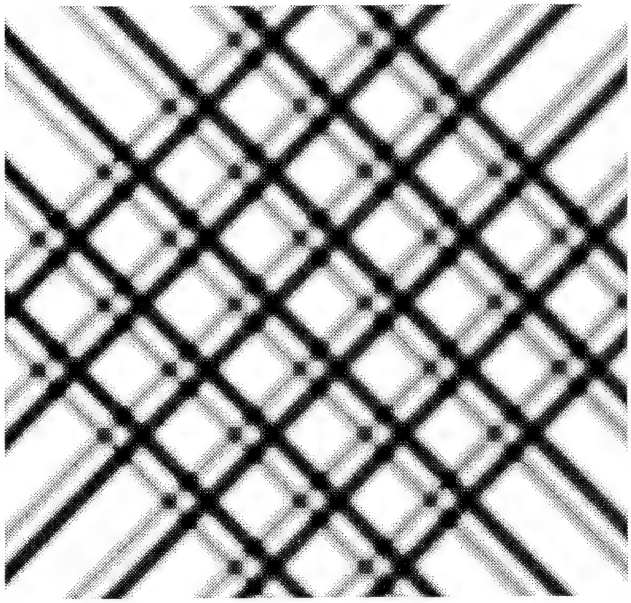
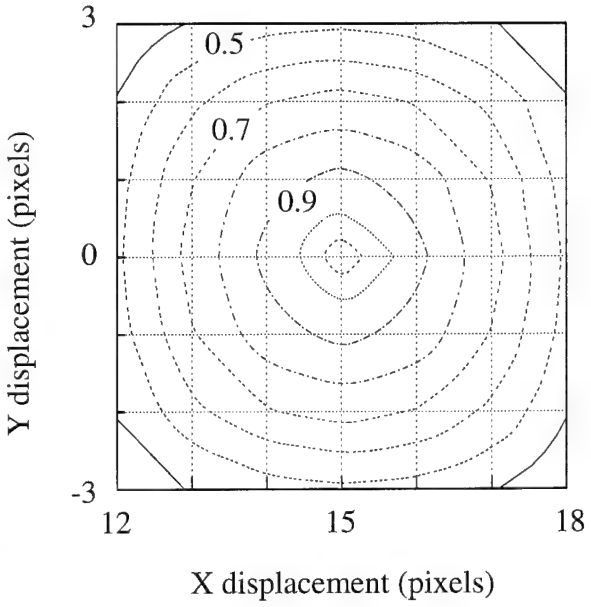


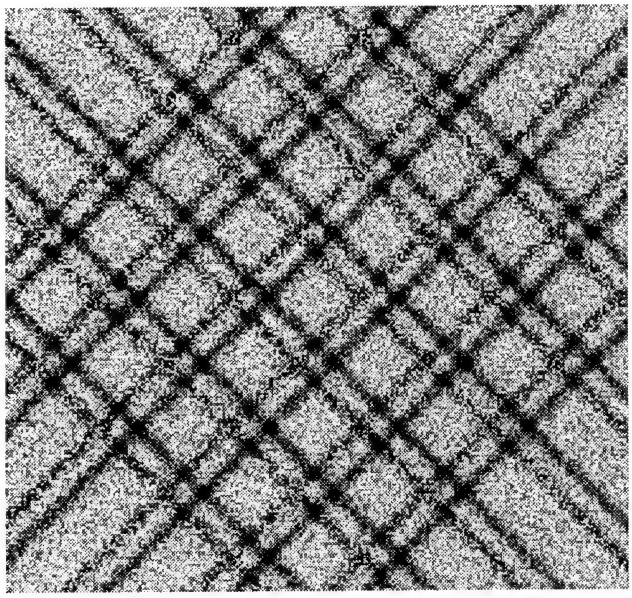
Figure 1. Optical and electronic arrangement for 2-camera MTV experiments. Both cameras view the same image plane through the cube beam splitter. Synchronization between the two cameras and laser is provided by a digital delay generator.



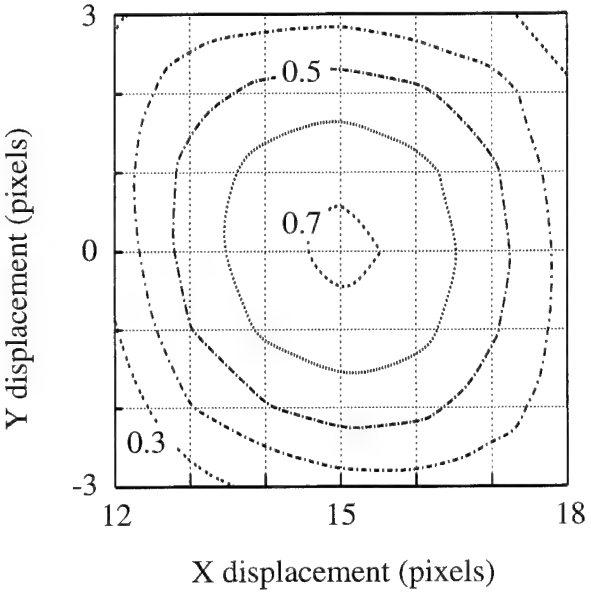
(a) No noise added



(b) Correlation coefficient for one intersection point in (a).



(c) S/N = 1.5



(d) Correlation coefficient for one intersection point in (c).

Figure 2. Simulated MTV grids and the corresponding correlation coefficient contours for one of the intersection points; Light lines: laser pulse at t= 0; Dark lines: MTV grid displaced by $\Delta x = 15$ pixels, $\Delta y = 0$ pixels. (a) and (b): No noise added. (c) and (d): S/N = 1.5.

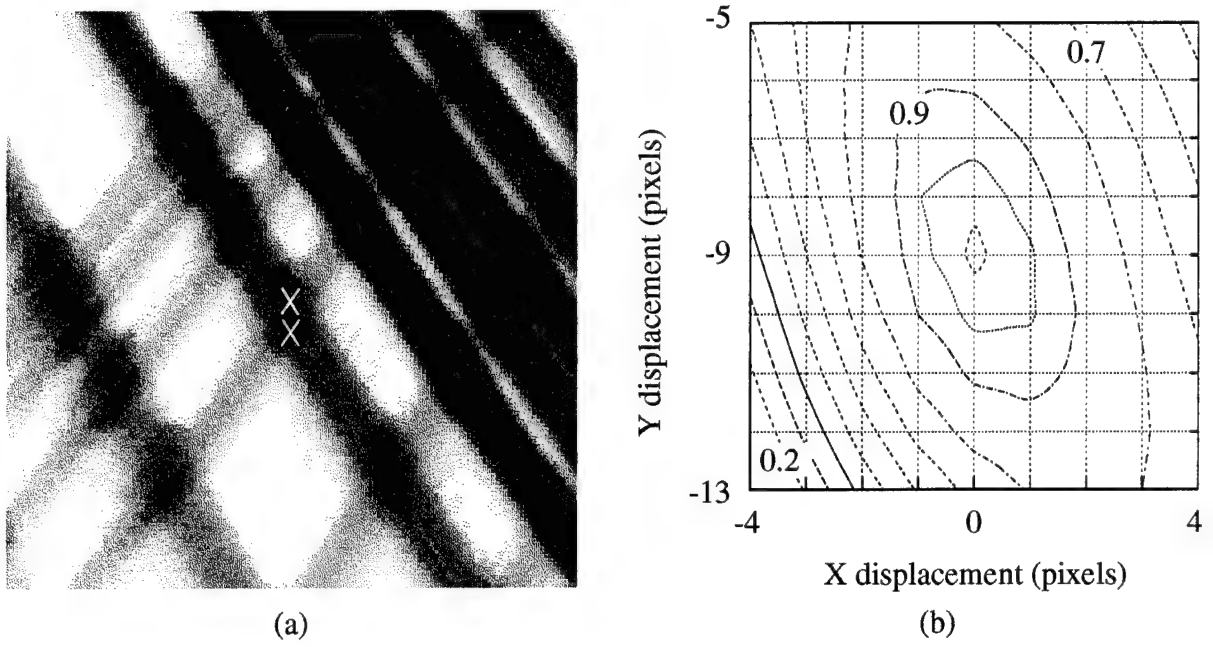


Figure 3. Experimental MTV grid. (a) Green lines are the grid at t = 0; red lines are the grid after a 4 msec delay. (b) Correlation coefficient contours for the indicated intersection in (a).

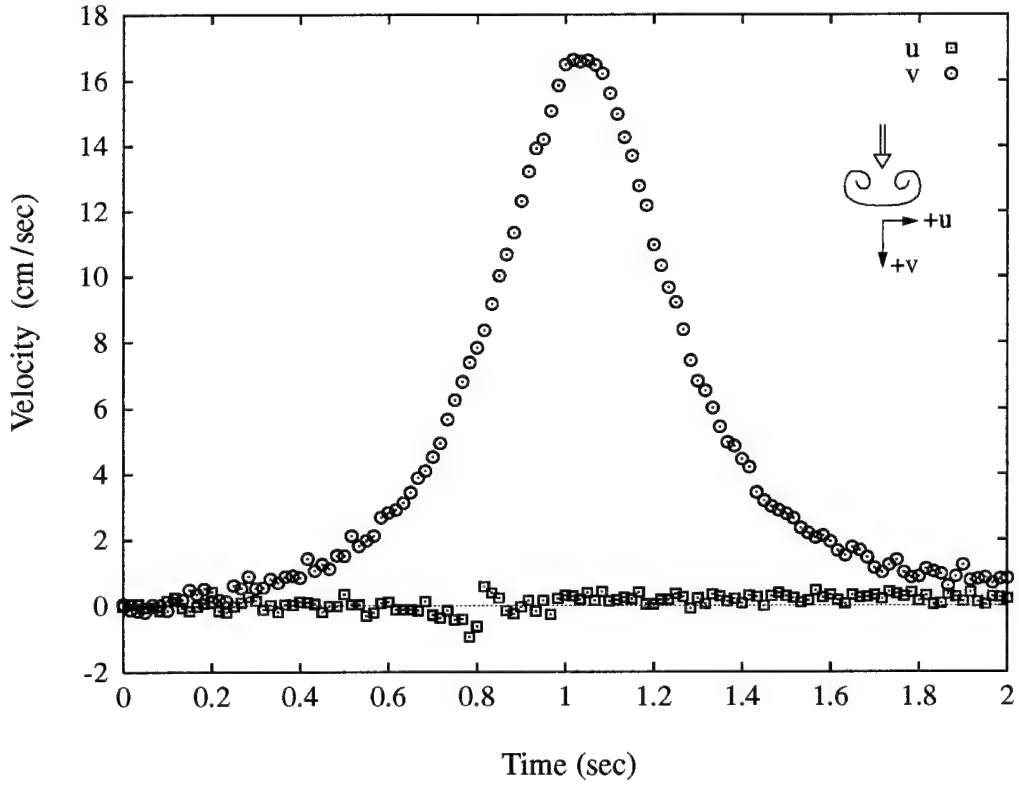


Figure 4. The time evolution of (u,v) velocity components measured by MTV near the center of a passing vortex ring.

(a) (b)

Figure 5. Instantaneous velocity vectors of the cross-stream flow in (a) an unforced and (b) a forced wake.

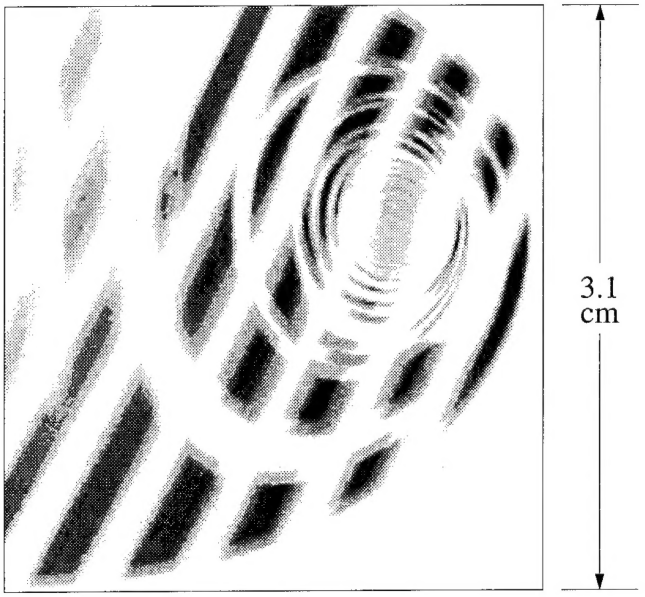


Figure 6. Lagrangian evolution of molecularly tagged fluid patches in a vortex ring. Only the right half of a downward-moving ring is shown. The lighter patches are regions imaged 20 μsec after laser firing; the darker patches are the same regions 9 msec later.

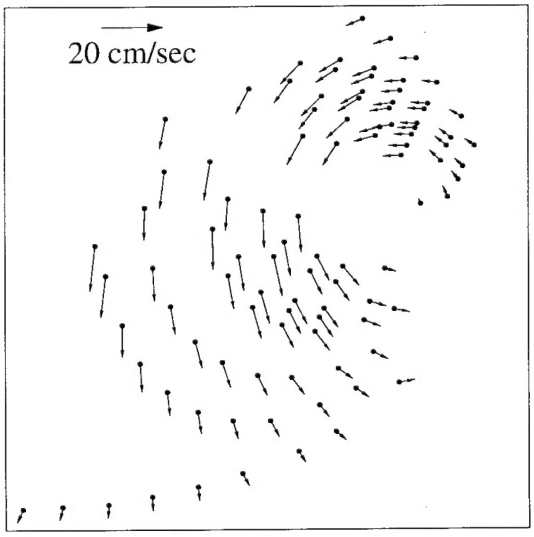


Figure 7. Velocity vectors inferred from Figure 6.

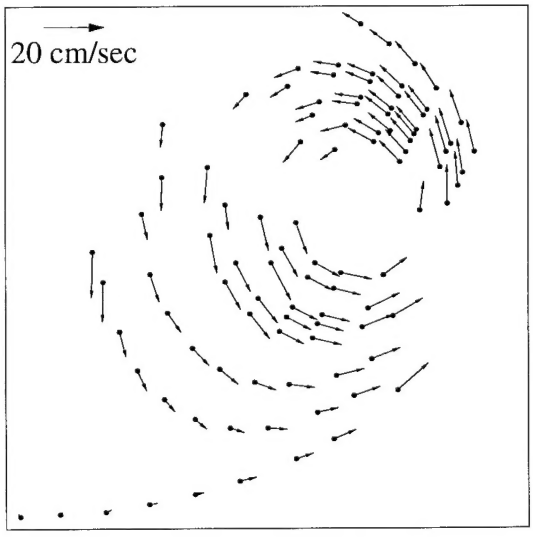


Figure 8. Velocity vectors inferred from Figure 6; the core convection velocity has been subtracted.

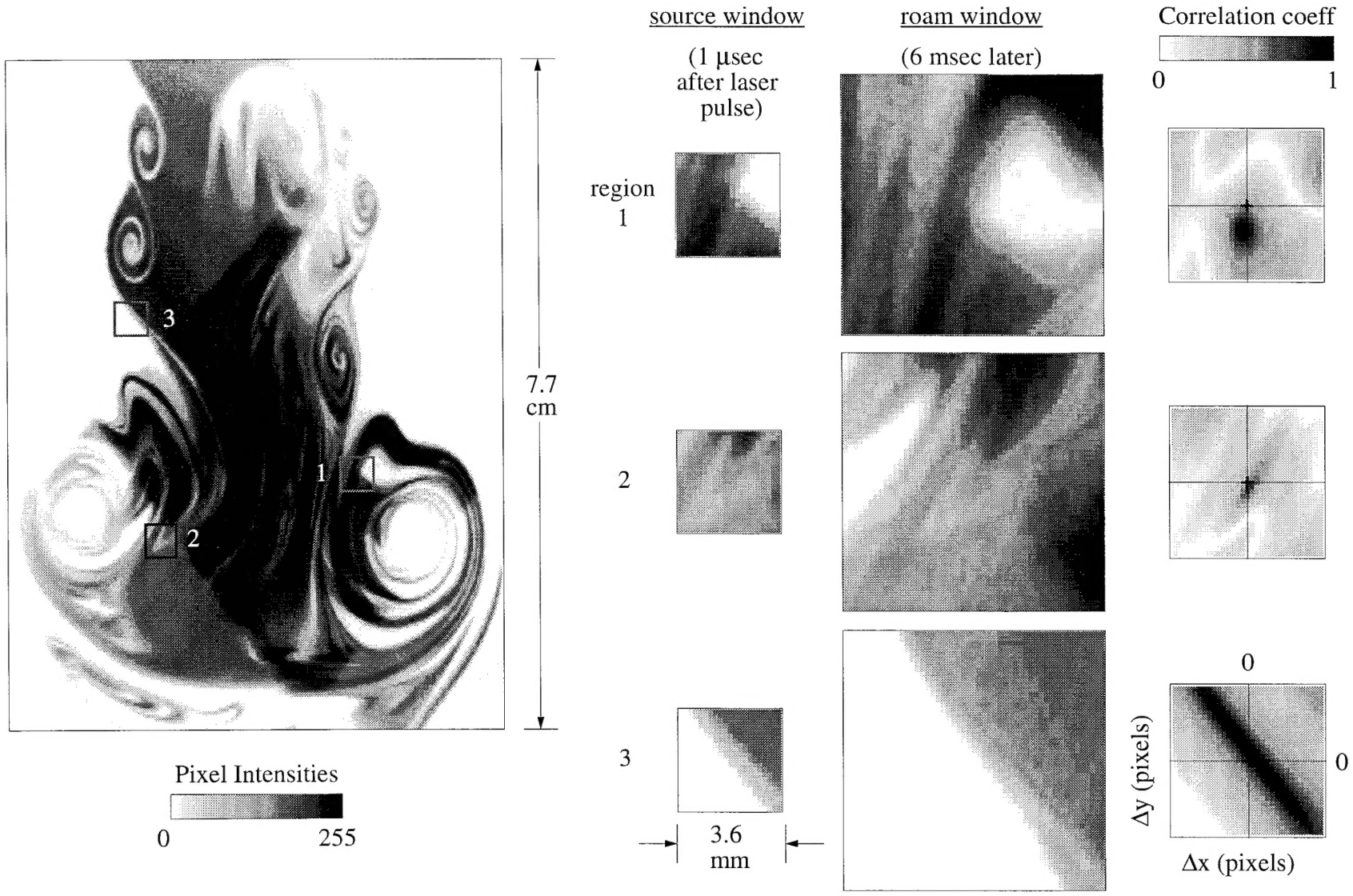


Figure 9. A molecularly tagged turbulent jet with non-uniform concentration field is shown on the left. The small images in the middle column show the pixel intensities of the tagged molecules from the 3 marked source regions at time $t=t_0$. The larger images in the next column show the tagged molecules within each roam window 6 msec later. The third column shows the resulting correlation coefficient fields. Velocities can be estimated accurately for regions 1 and 2, but not for region 3.

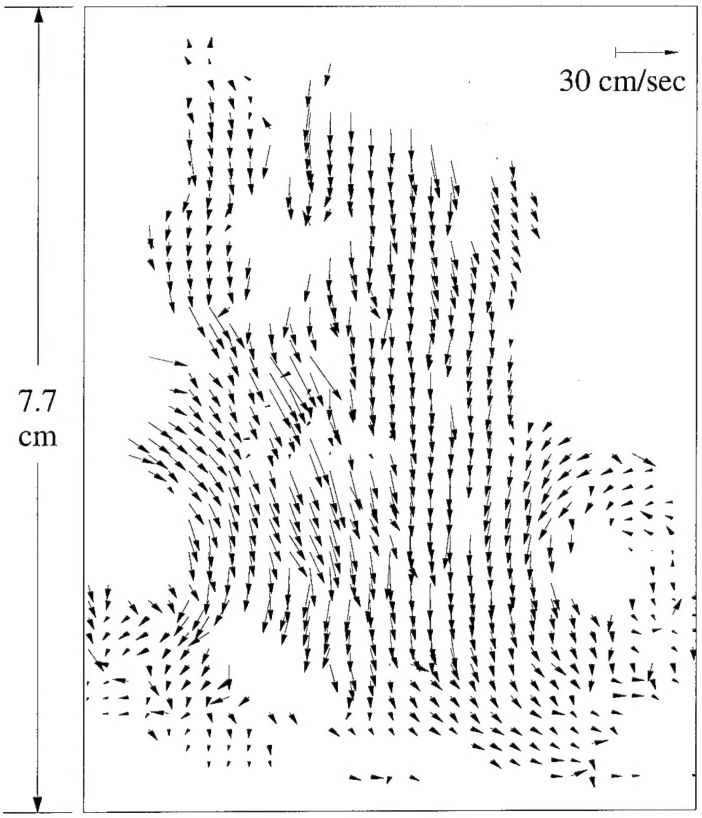
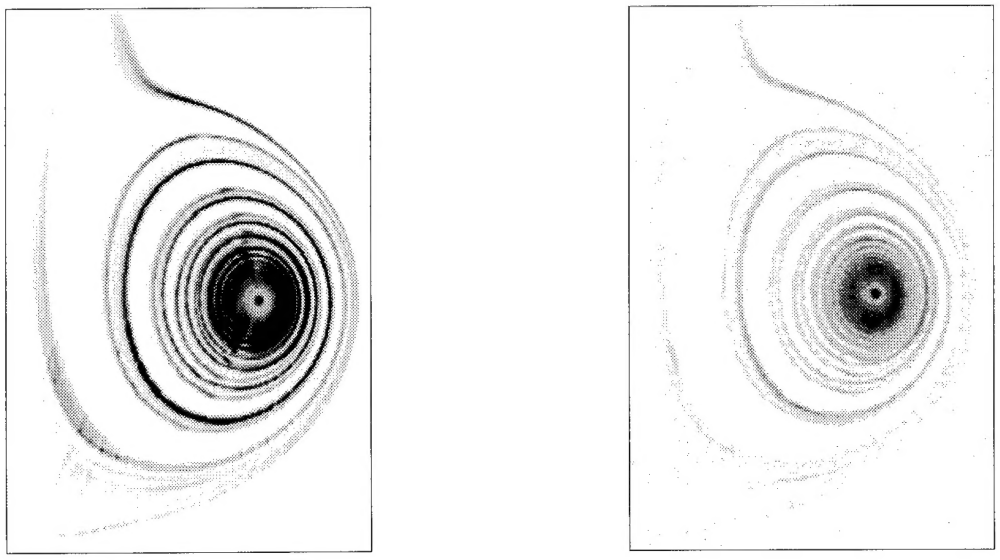


Figure 10. Resulting raw velocity vectors for the flow field illustrated in Figure 9.



(a) (b)

Figure 11. The chemically reacting interface between a vortex ring and the ambient fluid. (a) was imaged 20 μsec after tagging with a laser sheet; (b) shows the same interfaces 6 msec later.